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Significance of Regional Source Contributions to Urban PM-10 (Nominal 10 Micrometers) Concentrations

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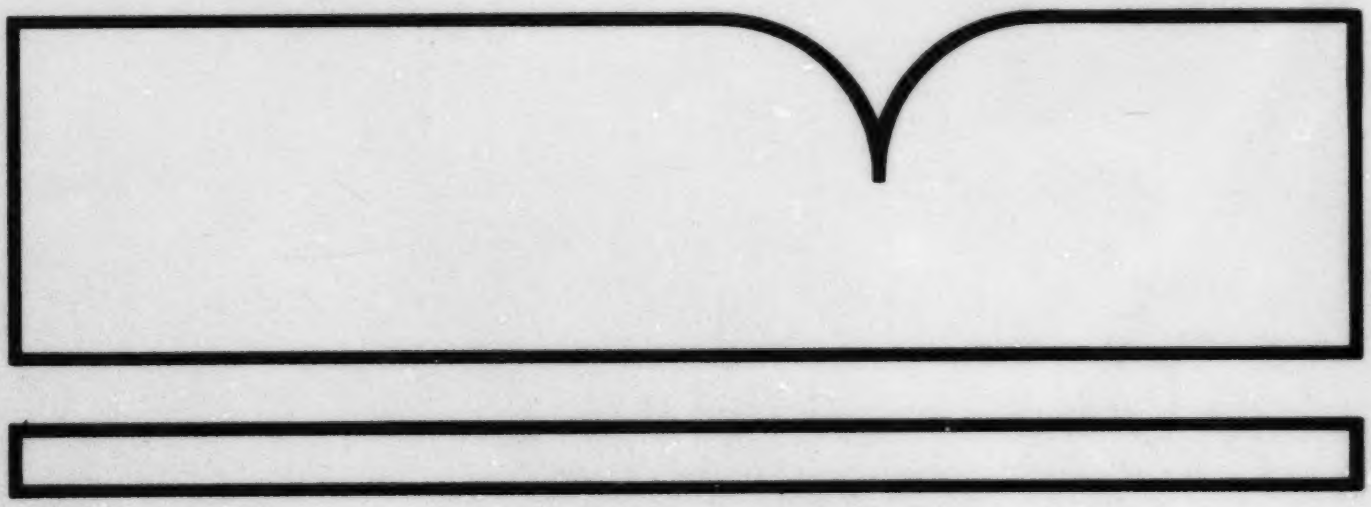

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SIGNIFICANCE OF REGIONAL SOURCE CONTRIBUTIONS TO URBAN PM-10 CONCENTRATIONS

by

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Abstract

Based on data collected in St. Louis, Philadelphia and other cities, we conclude that about 50-80% of the PM-10 concentrations is 'background.' The bulk of the background appears to be of regional origin. The chemical makeup of the background is sulfate as $SO_4^=$, 21-34%; crustal material, 14-36%; "unknown" (carbonaceous matter, ammonium, nitrate and water), 36-51%. Dispersion or receptor modeling using a local source inventory or elemental signatures can account only for the incremental contributions from local sources to the total PM-10 mass. It is argued that emission roll-back of local sources will have a limited effect on reducing total concentrations of PM-10.

INTRODUCTION

The U.S. Environmental Protection Agency has issued proposed revisions to National Ambient Air Quality Standards (NAAQS) for particulate matter.[1] The existing standards for total suspended particles (TSP) would be augmented by standards for particles with an aerodynamic diameter smaller than or equal to nominal 10 micrometers (PM-10). The proposed standards include probable ranges of the annual and daily averaged concentrations, the latter not to be exceeded more than once per year. Guidelines are given on monitoring methods and schedules but nothing is said in regard to what would be done if and when standards were exceeded. The apparent intention is to regulate PM-10 much in the same manner as TSP is regulated at present. Presumably, if standards were exceeded in an Air Quality Control Region (AQCR), emission roll-backs would be required of major contributing sources in that region under a state implementation plan (SIP).

There is a potential problem in regulating PM-10. Unlike TSP, sources contributing to the mass of PM-10 may lie outside a state as well as within. In some instances the out-of-state sources may have the predominant share. In such a case, an emission roll-back of identifiable local sources would have little effect on bringing the AQCR into compliance with standards, should these be exceeded.

The purpose of this paper is to show that in two eastern U.S. urban areas - St. Louis and Philadelphia - where extensive monitoring was undertaken, more than half of the PM-10 concentrations, both long-term (i.e. annual or seasonal) and short-term (daily) averages, is not of local origin, but is contributed largely by distant sources beyond the control of local authorities. A survey of the literature shows that other eastern U.S. urban areas may also experience large contributions to their PM-10 concentrations from non-local sources. This share of the PM-10 makeup is usually termed 'background.' Since the background constitutes such a large fraction of the PM-10 concentration, we argue that current monitoring, modeling and regulatory procedures that are used for TSP may not be suitable for PM-10.

THE ST. LOUIS STORY

In 1974-1977 a very extensive air quality monitoring study was performed in St. Louis (the RAPS study) by the U. S. EPA with particular attention to inhalable particles. Ten dichotomous samplers were deployed over an area of 40 x 90 km encompassing the St. Louis metropolitan area and rural surroundings. The dichotomous sampler inlets had particle size cutoffs at nominally 2.5μm (fine fraction) and 15μm (coarse fraction), respectively. The St. Louis coarse fraction includes somewhat larger particles than current dichotomous samplers which have a coarse fraction cutoff at 10μm. Indeed, some differences will be pointed out between the St. Louis measurements and those of subsequent studies.

The St. Louis measurements and data analyses have been described previously in the open literature[2-5], and EPA technical reports.[6-9] For easy reference, we reproduce in Figure 1 a map showing the location of the dichotomous samplers in the St. Louis area. For this analysis, we use measurements of one year, 1976.

The recommended procedures for estimating background concentrations use either the lowest average of concentrations at monitors, "excluding values when the source in question is impacting the monitor," or monitoring at a "regional"[10] site which is away from local sources, but impacted by similar sources. Because the northernmost (rural) monitoring site was not operating for much of 1976, we estimated background as the average of the two lowest concentrations found in the monitoring network in each (12 hour sampling) period. This is an operational definition of the background which is based on the available information. Such background estimates depend on the particular network used. Measurement errors and contributions by local sources to the lowest concentrations in the network may affect background estimates.

The annual average concentrations at the 10 RAPS sites of the PM-15, fine and coarse fractions are plotted in Figure 2. The background share averages 57.5% of the PM-15 at the 10 sites. The fine fraction background is 60%; the coarse fraction background is 55%. The ratio of average concentrations at the central industrial site (No. 1) to the most outlying site (No. 10) is only about 2 for PM-15, 1.75 for fine fraction, and 2.3 for coarse. There are two significant conclusions to be drawn from Figure 2. Concentrations at rural sites are due almost entirely to background particles and therefore receive little contribution from the metropolitan St. Louis sources. Equally important, the background concentration is a significant fraction of the total concentration at each site, even for the central city sites.

To elaborate this point further, in Figure 3 we plot the daily concentrations at the 10 sites for all days for which data are available in 1976 vs. the background concentrations on that day. For any background concentration, the vertical spread of the points indicates the range of the 24-hourly concentrations at the individual sites. The lower envelope line projects the background concentration onto the ordinate. In the left half of the figure, where concentrations are low, the spread between individual sites can be several times the background concentration. However, on high pollution days (right half of figure), the ratio of the highest to lowest concentrations rarely exceeds 2. The interesting conclusion is that on low pollution days local sources may contribute relatively more to the total concentrations. On high pollution days (when standards are likely to be exceeded), local impacts become less pronounced and most particles seem to come from outside the network.

There is another indication that the bulk of the particles does not originate from local sources. In Figure 4 we show the annual average pollution roses for fine and coarse particles at 10 RAPS sites. The roses are divided into 16 arms corresponding to 22.5° wind sectors. Each arm length is proportional to the average concentration of those samples in a year that have been obtained when the resultant wind for the sampling period (12 hours) blew from that sector.[5] While the inner metropolitan area sites Nos. 1-5 generally have long arms, the arm lengths are nearly uniform around the compass and do not point to any particular local point emission source. In the fine fraction one notices slight arm elongation toward the east-southeast. Since there are no identifiable local sources to the east, the arms evidently point to some distant sources, most likely coal-using sources to the east. Indeed, the sulfur pollution roses which we have shown previously[5] point in the same direction. As described below, sulfates constitute the bulk of the fine fraction.

The annual average chemical makeup by weight, of PM-15 is depicted in a pie chart (Figure 5). The chemical analysis of the samples is based on X-ray fluorescence,[9] which measures elemental concentrations. The pie chart is based on the assumption that the elements are present in the aerosol as their oxides, e.g. $SO_4^=$, SiO_2, Al_2O_3, CaO, etc. Of the PM-15 mass, 21% is $SO_4^=$, 39% is 'crustate' ($SiO_2 + Al_2O_3 + Fe_2O_3 + CaO$), and 35% is 'unknown.' The latter includes organic and elemental carbon, ammonium, nitrate, crystalline and ab-ad-sorbed water. Part of the ammonium and water could be associated with sulfate. Indeed, a regression analysis[11] indicates that the mass associated with sulfur in St. Louis is 5.1 times the sulfur mass, rather than 3 expected stoichiometrically for $SO_4^=$. Using this higher value, the sulfate associated mass would be 36% of the total.

We also determined the chemical composition of the fine and coarse fractions separately. In the fine fraction, sulfate (as $SO_4^=$) comprises 42%; in the coarse fraction, crustate comprises 68%. The latter high percentage may be due in part to the high cutoff (15 μm) of the RAPS dichotomous samplers allowing the collection of wind blown dust. The chemical composition of the 24-hour samples, including peak concentrations, is not greatly different from the annual averages.

We reported previously[5] on dispersion modeling of sulfate. Using the St. Louis emission inventory of primary sulfate and reasonable transformation rates of SO_2 to $SO_4^=$ (5% per hour during the day; 2% per hour at night) we could account for only 10-15% of the observed sulfate levels. The rest evidently comes from distant sources. Fay et al.[12] used a long range transport model to apportion the St. Louis annual average sulfate concentration. It was found that the following states were the major contributors: Missouri (11.9%), Indiana (10.3%), Texas (8.9%), Ohio (7.6%), Illinois (6.7%), Tennessee (6.6%), Kentucky (6.3%), and Alabama (5.1%). Part of the crustate may also come from distant stationary combustion sources. For the Washington, D.C. aerosol, Kowalczyk[13] estimated that about 30% of the Al and 80% of the Fe is coal related (Si was not determined).

THE PHILADELPHIA STORY

The Philadelphia Area Field Study (PAFS) was conducted by EPA for four weeks in July and August, 1982. The Philadelphia urban area is considerably larger than St. Louis, yet the monitoring network is smaller. Six dichotomous samplers were deployed over an area of 20 x 30 km (Figure 6). The coarse fraction cutoff was at a nominal 10 μm, in line with the proposed reference

methods.[1]

The monthly average concentrations at the 6 PAFS sites of total, fine and coarse fractions are plotted in Figure 7. Clarksboro was the most rural of the sites, probably least impacted by local sources. Fireboat was deployed near ongoing construction activity; this site gave peculiar results. In the following analysis, site-averaged concentrations are computed for 5 sites only, excluding Fireboat.

The background concentration was estimated using the lowest observation in the network at each sampling time. The background share of the average PM-10 concentration is 77%. The fine fraction background is 83%; the coarse 64%. The ratio of concentrations at the center city site Broad Street to that at Clarksboro is less than 1.2 for total mass and fine fraction, and 1.6 for the coarse fraction. Thus, in Philadelphia the background share is even larger than in St. Louis, and the relative difference between city center and outskirts is smaller. These differences between Philadelphia and St. Louis may be due in part to the smaller size cutoff of the coarse fraction inlet and to the difference between summer and annual averages. In addition, the smaller monitoring network in Philadelphia may tend to overstate the background fraction, due to possible contributions from local sources received at all monitoring sites.

The daily concentrations are plotted in Figure 8 in which the letter symbols stand for the 6 sites. The envelope line equals the background concentration; the spread indicates the range of concentrations at the sites. Excluding the Fireboat site (F), we see that the range is usually less than twofold. As in St. Louis, the range is greater at lower overall concentrations. This again indicates that the proportionate contribution of local sources is less at high concentrations when exceedances might occur.

The chemical composition (monthly average) is depicted in the pie chart of Figure 9. The $SO_4^=$ component is 34%, compared to St. Louis' 21%. Regression analysis assigned a multiplier of 4.2 to Philadelphia sulfur; thus the total sulfur-related contribution is more likely to be 48%. The crustal portion is 14% vs. St. Louis' 36%, the 'unknown' is 51% vs. St. Louis's 36%. These differences may be caused by several factors: actual chemical makeup differences, annual vs. monthly averaging, and different dichot samplers.

TALE OF OTHER CITIES

Although the Inhalable Particle Network (IPN) has been operating for several years,[14] few of the available studies allow the determination of background levels in urban areas, and the origin of the background. Wolff et al.[15] deployed 4 dichotomous samplers in and around Detroit, MI, in summer 1981. Sulfate constituted 40-50% of the fine fraction. There were no $SO_4^=$ differences among the sites. In Detroit, only 18% of the coarse fraction was due to crustate, of indeterminate origin. The most important local influence was observed at Dearborn, at the edge of a heavily industrialized area, but even at this site local influences appeared to be smaller than the regional ones. Jordan and Beranek[16] operated 4 dichot monitors in and around Indianapolis, IN, for one year in 1984. They noticed a lack of differences in PM-10 concentrations at the 3 urban-industrial sites (the 4th was southwest of the city), and attributed it to "a lack of significant PM-10 sources in the vicinity of monitors." The ratio of annual concentrations of PM-10 between a center-city site and the background site was 1.2. Shaw and Paur[17] analyzed the data collected from 1979 to 1981 by 3 dichot samplers deployed in rural areas along a 590 km arc in the Ohio River Valley. The sites showed a high

correlation of total, fine and coarse fraction mass, and elemental composition at all sites, indicating the impact of regional, rather than local sources on the widely scattered samplers. We note that the PM-10 concentrations at these sites were quite similar to the background in St. Louis and Philadelphia.

BACKGROUND AND PM-10 REGULATIONS

Since background is such a large fraction of the PM-10 mass, we see several problems in regard to the proposed PM-10 regulations. These problems lie in the areas of monitoring, modeling and enforcement.

Monitoring. The proposed revisions of NAAQS to include PM-10 give little guidance regarding the determination of background in an AQCR. The traditional "upwind" or "regional" site may not necessarily represent the true background. We believe that it may be necessary to run 6-10 dichotomous samplers continuously for a year or more within an AQCR to obtain reliable statistics of local and regional (background) contributions.

Modeling. Traditional approaches to TSP apportionment are based on dispersion and receptor modeling. For PM-10, these approaches may work only for the local increment, over and above the background concentration. However, even for the local increment, it will be required to have accurate PM-10 source inventories and elemental signatures -- which at present are scarcely developed.

Enforcement. In the above case studies, at a few monitoring sites the PM-10 mass approached or exceeded the proposed short-term standard. The background in those instances amounted to more than half of the observed concentrations. In cases where a significant exceedance is encountered, it will be difficult to bring these areas into attainment of standards by rolling back emissions from local sources only.

CONCLUSIONS

From several studies in the eastern U.S. one may draw the following conclusions:

o There are relatively small differences in PM-10 concentrations among widely dispersed sites throughout an urban airshed. This indicates the lack of impact of local sources, and conversely, the significance of regional sources on urban PM-10 concentrations.

o The background concentration of PM-15(10) in St. Louis and Philadelphia, amount to 50-80% of the total.

o The primary chemical constituent of the background concentration is sulfate with associated cations and water of hydration. The bulk of sulfate must originate from distant sources because local source inventories of primary sulfate plus build-up of secondary sulfate can account for only a small fraction of the observed concentration.

o The second largest component of the background is crustal material (SiO_2, Al_2O_3, CaO, Fe_2O_3). Since the elemental composition of flyash from large stationary combustion sources is similar to that of fugitive dust from soil comminution, it is difficult to apportion the crustal matter to these sources. One study[13] assigned more than 30% of the crustal matter to coal combustion.

One may also draw several inferences in regard to PM-10 regulatory issues:

o The determination of true background concentrations may require the use of 6-10 monitoring sites over a year or more.

o Dispersion modeling of PM-10 based on local source inventories can account only for the local increment of PM-10.

o Receptor modeling cannot distinguish between local and regional sources having comparable signatures, and hence does not distinguish the origin of the bulk of PM-10, sulfate and 'crustate.'

o If exceedances of standards (either short- or long-term) were to occur, roll-back of local emissions may not be sufficient to bring an air quality control region into attainment of standards.

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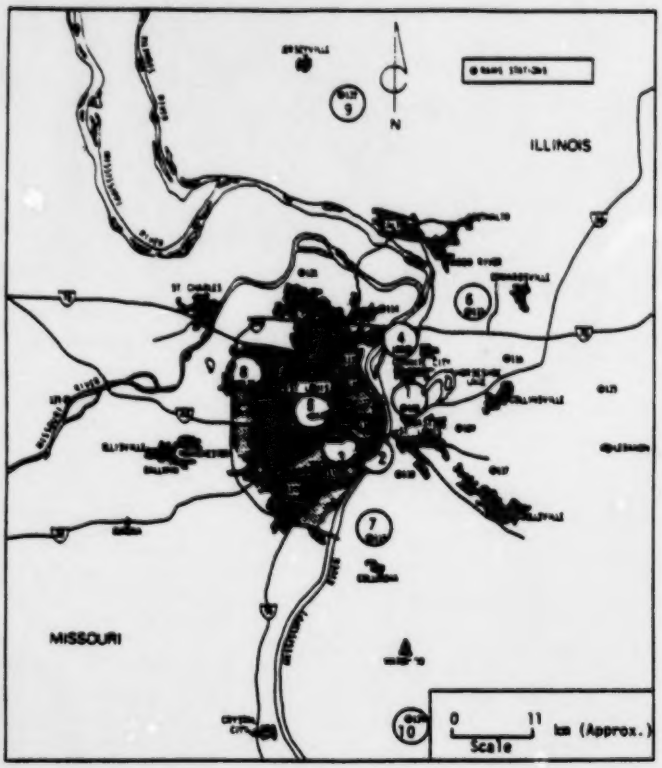

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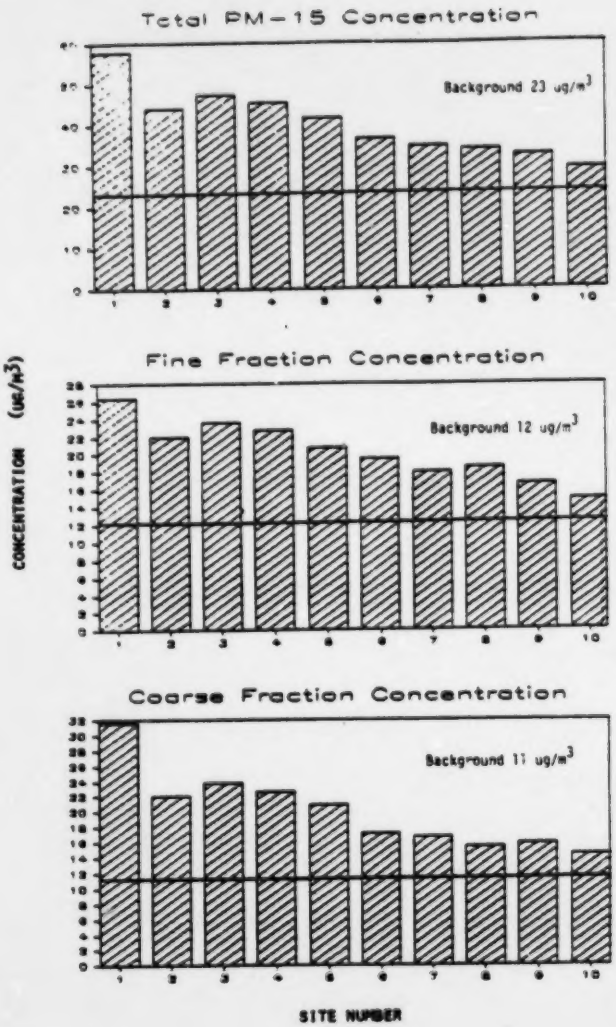

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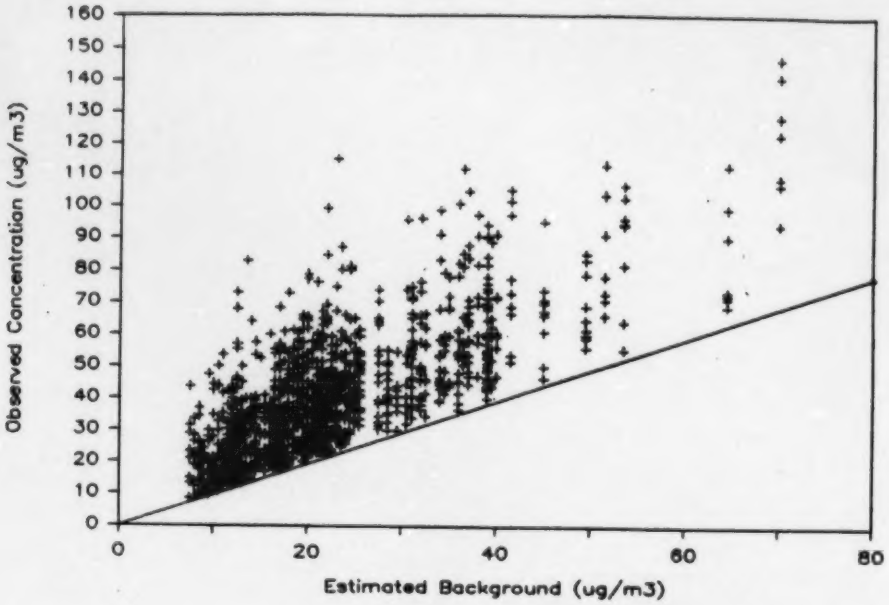

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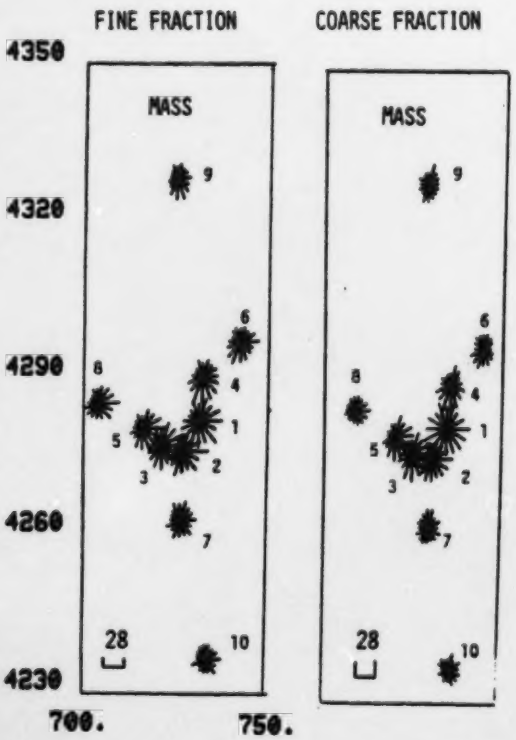

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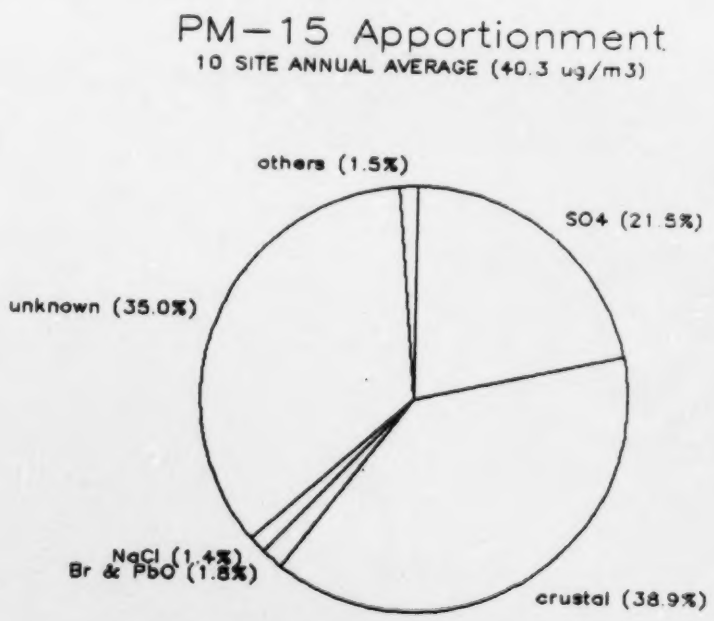

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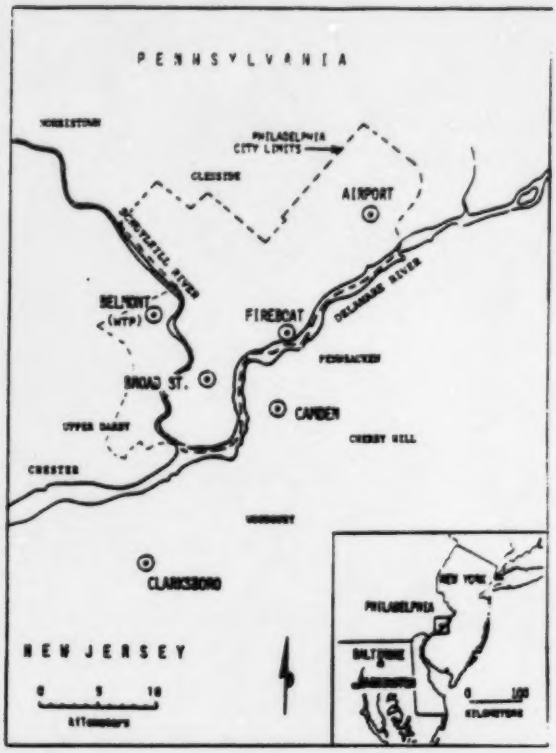

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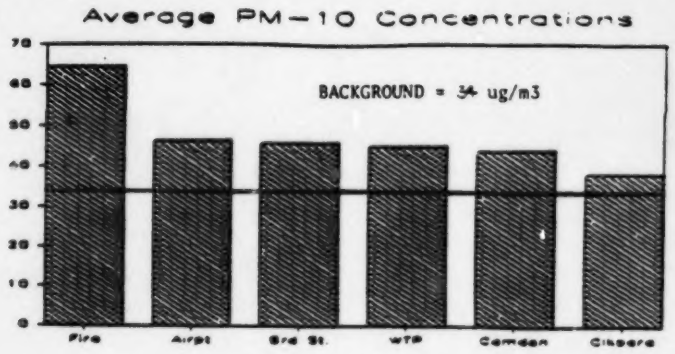
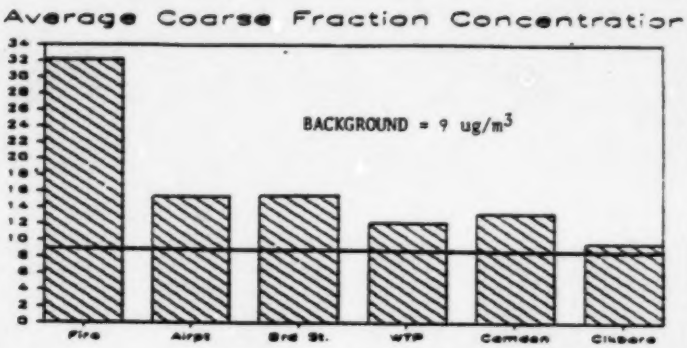
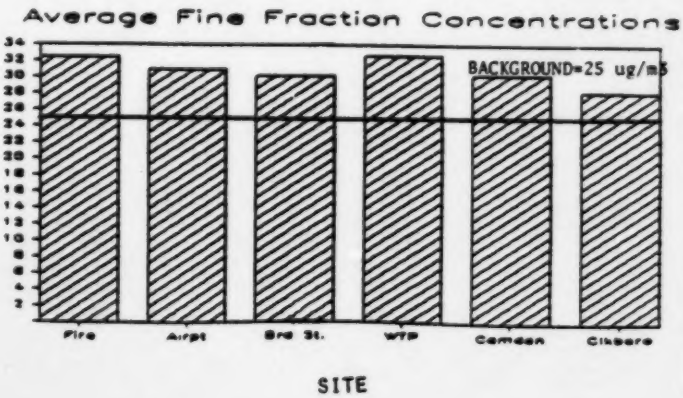
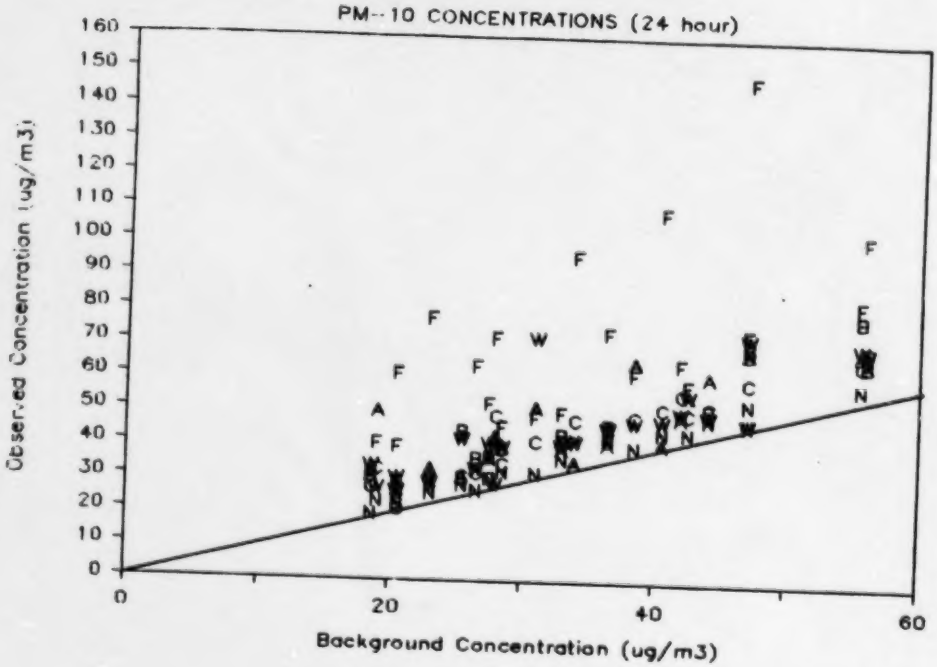


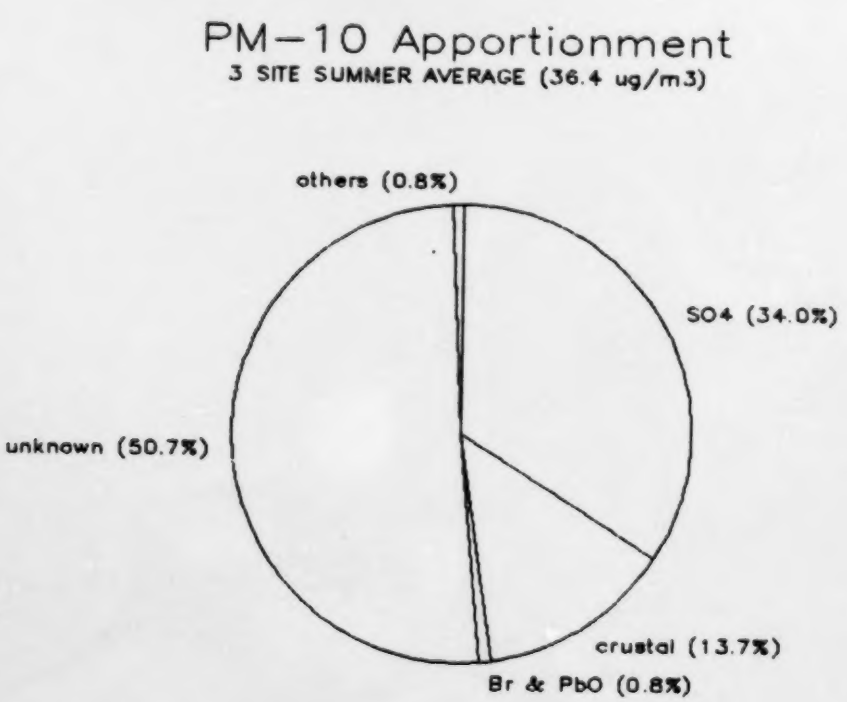




7. Monthly (Jul/Aug, 1982) average concentrations at 6 sites in Philadelph (a) Total PM-10; (b) fine fraction; (c) coarse fraction. Background indicated by horizontal line (average of lowest concentration in networ in each 12 hour sampling period).



8. Total 24-hour PM-10 concentrations in Philadelphia in July/August 1982 at 6 sites (letter symbols) vs. estimated background concentration (lower envelope line). Vertical spread of data indicates concentration range at 6 sites.



9. Chemical composition of Philadelphia PM-10. Monthly, 6 site average.